

REMARKS/ARGUMENTS

Reconsideration and allowance of the present application based on the following remarks are respectfully requested.

Upon entry of the above amendments, claims 14, 20-21 (withdrawn) and new claims 30-49 will be pending. Of the new claims 30-49, claim 30 is independent and generic to the embodiments as set forth in claims 1-3, and 22. New claim 31 substantially corresponds to prior claim 3 (one pot reaction) while new claim 32 substantially corresponds to prior claim 2 (two-step reaction). Claims 33-35 substantially correspond to prior claims 25, 23 and 24, respectively. New claims 36-41, 46 and 48 correspond to previously presented and withdrawn claims to chemically surface-modified silica gels. New claims 42-45, 47 and 49 correspond to previously presented and withdrawn claims to various uses of chemically surface-modified silica gels.

Accordingly, no new matter is added by the newly presented claims.

In view of the presentation of the new independent claim 30, which encompasses the one-pot reaction and the two-step reaction of previous claims 1-3, it is respectfully requested that the restriction requirement be reconsidered and withdrawn.

The following discussion is presented by way of background in order to facilitate an understanding of the subject matter encompassed by the disclosure and the pending claims and the differences between Applicants' invention, as claimed, and the prior art.

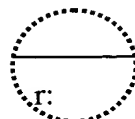
In order to modify a nanopore silica gel with a high loading of ligands, the gel should have a plurality of open channels within the gel structure and silanol (Si-OH) groups on the surface. A freshly prepared wet silica gel normally meets these requirements. At the completion of a gelation reaction, only two of the four silanol groups of each silica atom are reacted to form a Cayley tree structure. There are two remaining silanol groups of each silica atom which could be used for surface modification. However during the subsequent drying and aging, a silica gel could easily lose these remaining surface silanol groups through intrapore condensation reactions that could permanently close off the open channel structure at the same time.

Several material factors dictate the intrapore condensation and crosslinking reactions. First a liquid-air meniscus formed within the pores during drying exerts a strong capillary stress due to interfacial tension. This surface stress is directly proportional to the surface

tension and inversely proportional to the pore size. For nanometer size pores, the stress can be in the range of 100 Mega Pascal.

$$\text{Capillary stress} = \frac{\text{surface force}}{\text{surface area}} \approx \frac{\sigma \cdot 2\pi r}{4\pi r^2} = \frac{\sigma}{2r}$$

σ : surface tension



Secondly, the liquid-air mixture in the pore has an infinite compressibility and would provide no support to oppose such high surface stress. The pore shrinkage caused by the combination of high surface stress and infinite compressibility promotes the crosslinking reaction among surface silanols. The condensation of the surface silanol groups reduces the amount of surface silanol groups available for modification reaction and permanently closes off the originally open channel structures.

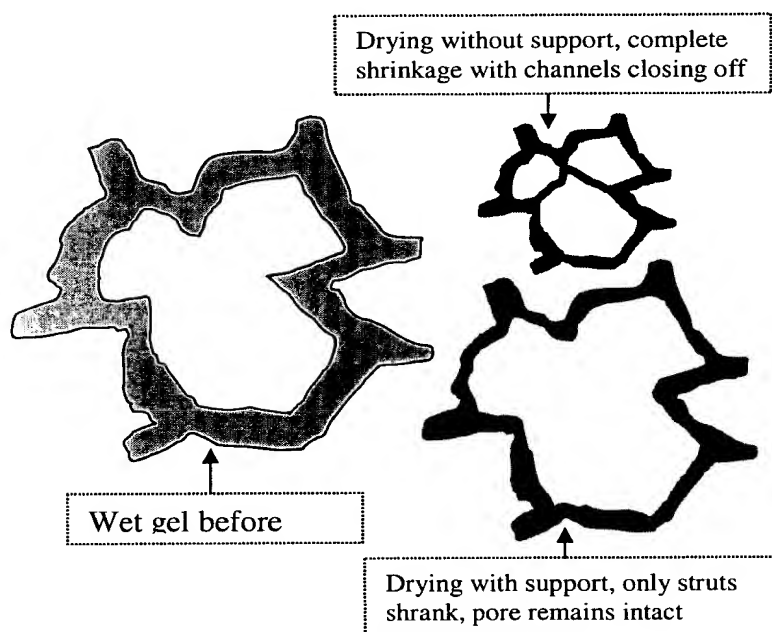
Since the days of making silica aerogels (1980), there have been a series of processing steps designed to prevent shrinkage during the removal of solvent from a silica gel. Most noticeable are the following three approaches:

- (1) Using solvent exchange to replace water with an organic solvent which has a much lower surface tension and therefore resulting in a much lower surface stress.
- (2) Apply drying under a supercritical condition so that there is no surface stress and liquid meniscus during the drying process.
- (3) Reacting remaining surface silanol groups to reduce its moisture sensitivity and preventing subsequent pore shrinkage caused by adsorbed moisture.

In 1992, there was a technology breakthrough in this area developed by scientists at Mobil. A family of mesoporous molecular sieves (M41S) was synthesized [J. S. Beck, et al; *J. Am. Chem. Soc.* 1992, 114, 10384 (1992); C. T. Kresge, et al, *Nature* 359, 710 (1992); J. S. Beck, et al, *Chem. Mater.* 6, 1816 (1994)] by using cationic surfactants to assemble silicate anions from solution. The micellar assemblies of quaternary ammonium cations served as the structure-directing agents. Their strong electrostatic interactions with anionic silicate oligomers led to condensation of inorganic precursors around the regular structure, forming a continuous silica phase with templated pore morphology.

The Mobil technology provided the basis for a promising synthesis process that could create and maintain a high amount of mesopores with minimum shrinkage. It utilized

surfactant self-assembly to create a desired morphology and then relied on the same surfactants to support the pore structure against shrinkage during aging and drying. If this scheme is used along with adequate aging, the condensation of silanol groups will occur primarily in the strut, not across pores. This will restrict the shrinkage primarily to the strut areas, making the backbones stronger, while leaving the vast pore volume unaffected by drying. This phenomenon is illustrated below.



However, this technology still suffers the loss of surface silanol groups due to crosslinking within struts during drying and aging. Once the ideal pore morphology is created, there are very few silanol groups left for surface modification by ligand groups. This phenomenon is identical to a dried and shrunken silica gel except the channel structure is considerably better than regularly dried gel. To deal with the loss of surface silanol groups prior to surface modification reaction, one must rehydrolyze the silica surface to create more silanol groups. This turns out to be a more difficult task because a hydrolysis reaction in ample water could destroy the strut and pore structure as well. Thus, a common practice is to use a large amount of organic solvent and a controlled amount of water for hydrolysis of crosslinked silica surface. This type of hydrolysis would require an extended period of reaction time under refluxing with excess amount of organic solvent.

To summarize, the prior art may be characterized by one or more of the following features which individually or collectively serve to differentiate these techniques from the Applicants' claimed methodology and end product:

- (a) Using a low surface tension (normally organic) solvent to exchange with water. The amount of organic solvent is normally several times of the initial water used. Phase separation occurs.
- (b) Surface modification is done after the solvent exchange with or without refluxing depending on whether there were enough surface silanol left in the gel.
- (c) Supercritically or freeze drying the gel to reduce the shrinkage and crosslinking, followed by surface modification with ligands. The open channels may be preserved, but at the expense of the concentration of the surface silanol groups. Consequently, the achievable ligand loading is much lower than in Applicants' method.
- (d) Mobil template technology followed by a hydrolysis reaction to regenerate the surface silanol groups; normally requires refluxing with water-containing organic solvent for an extended time before a surface modification reaction.

With the above background in mind, it is respectfully submitted that the methods of making, products made and methods using such products, as set forth in the rejected and pending claims may be readily distinguished from and, would not have been obvious in view of, the cited prior art.

Claims 1, 13, 14, 22, 25 26 and 29 were rejected under 35 U.S.C. 102(b) as anticipated by, or under 35 U.S.C. 103(a), as unpatentably obvious over, Burns et al, U.S. 5,708,069.

This rejection is respectfully traversed for at least the following reasons.

Since the feature from step (c) of claim 2 is included in new claim 30, this rejection is no longer applicable to any of the pending claims and should, therefore, be withdrawn.

Moreover, with respect to the subject matter of claim 14, the disclosure of Burns et al does not include a step of selecting a ligand molecule for the target specie by determining bond energy between the second functional group and target specie or by determining

solubility product constant, K_{sp} . In fact, Burns et al does not at all address preparing a chemically surface modified silica gel effective for adsorbing a target specie but, rather, is concerned with silica gels as filler materials.

Accordingly, reconsideration and withdrawal of the rejection based on Burns et al is respectfully requested.

Claims 1, 2, 13, 14, 22-26 and 29 are rejected under 35 U.S.C. 102(b), as anticipated by, or under 35 U.S.C. 103(a), as unpatentably obvious, over Lentz, U.S. 3,122,520.

This rejection is respectfully traversed for at least the following reasons.

The process disclosed by Lentz does not include a step of creating and preserving a plurality of open channels within a gel structure and a plurality of surface silanol groups by preventing crosslinking condensation from occurring, either generally or specifically, prior to reaction with a ligand-carrying silane coupling reaction.

In particular, Lentz discloses a method for forming silicone rubber fillers by heating a silica hydrosol under strong acid conditions and subsequently forming an organosol by solvent exchange, using, *e.g.*, hexamethyldisiloxane, often as both reactant and solvent, simultaneously or with solvent exchange with, *e.g.*, hexane or toluene. As noted from the Examples, the quantity of the organic solvent was several times higher than the amount of water being exchanged. This process, therefore, necessarily results in phase separation while not creating and preserving a plurality of open channels within the gel structure by preventing crosslinking condensation from occurring.

Therefore, notwithstanding the step of cooling the mixture of the acid refluxed hydrosol and isopropyl alcohol (used as a water-miscible solvent), to 70°C (Example 7) or the step of heating the strong acid refluxed hydrosol to 80°C (Example 9), these processes do not preserve the open-channel structure or prevent crosslinking and do not result in reacting substantially all of the surface silanol groups with the reactive ligand coupling reagent.

Moreover, with regard to the one-pot reaction as set forth in claim 31, the process of Lentz first forms a silica hydrogel by the strong acid heating step followed by reaction with the silane reactant. With regard to the two-step reaction as set forth in claim 32, the process of Lentz does not maintain a freshly prepared silica gel at a temperature in the range of from 40 to 80°C. for from 30 to 60 minutes. In Example 7 of Lentz, the hydrogel was refluxed for 3 hours and in Example 9, the hydrosol is heated for 24 hours.

With regard to claim 14, the disclosure of Lentz et al does not teach this subject matter for at least the same reasons as set forth above with respect to the Burns et al patent.

Accordingly, it is respectfully submitted that the process and product disclosed by Lentz does not anticipate or render obvious the subject matter as set forth in the pending claims.

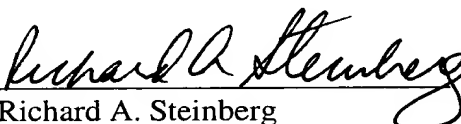
Therefore, reconsideration and withdrawal of the rejection based on Lentz is respectfully requested.

Therefore, all objections and rejections having been addressed, it is respectfully submitted that the present application is in condition for allowance and a Notice to that effect is earnestly solicited.

Should any issues remain unresolved, the Examiner is encouraged to contact the undersigned attorney for Applicants at the telephone number indicated below in order to expeditiously resolve any remaining issues.

Respectfully submitted,

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